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Review

State of the Art and Environmental Aspects of Plant Microbial Fuel Cells' Application

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Abstract: Environmental pollution is becoming ubiquitous; it has a negative impact on ecosystem diversity and worsens the quality of human life. This review discusses the possibility of applying the plant microbial fuel cells (PMFCs) technology for concurrent processes of electricity generation and the purification of water and soil ecosystems from organic pollutants, particularly from synthetic surfactants and heavy metals. The review describes PMFCs' functioning mechanisms and highlights the issues of PMFCs' environmental application. Generally, this work summarizes different approaches to PMFC development and to the potential usage of such hybrid bioelectrochemical systems for environmental protection.

Keywords: plant microbial fuel cell; electrogenic microorganisms; biofuel cells; synthetic surfactants; ecosystem cleaning; reduction of greenhouse gases; environmental protection

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1. Introduction

The ubiquitous environmental pollution due to various anthropogenic substances, such as heavy metals [1], petroleum products [2], medicinal preparations [3], and pesticides [4], is one of the main problems of mankind nowadays. Pollutants have a negative impact not only on the environment but also on human life, accumulating in heterotrophic food chains and entering the human body, which leads to various diseases of the nervous system and respiratory organs, as well as genetic abnormalities while reducing life expectancy [1]. The above-described problems are reflected in the UN Sustainable Development Goals; according to the developed programs of the United Nations Environment Programme (UNEP) and UN-Water, the control over the global pollution of ecosystems and their restoration are of high priority [5].

Bioremediation, a complex of purification methods using the metabolic potential of biological objects, is applied to purify soil and water ecosystems from pollutants. Thus, the introduction of microorganisms into ecosystems makes it possible to dispose of various pollutants by converting them to simpler safe substances. The principle of phytoremediation is based on the binding and accumulation of pollutants in plant vacuoles [6]; it activates a complex metabolic pathway involving the antioxidant plant protection system [7]. Additionally, plants and microorganisms (fungi and bacteria) interact with each other at the root level (in the rhizosphere), showing a positive synergistic effect in the elimination of pollutants such as heavy metals and organic compounds [8,9]. The disadvantages of the bioremediation of soils include the low rate of toxicant biodegradation, as well as the need for a thorough preliminary examination of the contaminated site to clarify the modes of biotechnological work. This requires high labor and energy costs, such as the plowing and irrigation of fields and the disposal of waste plants. Therefore, this technology is not widely used in developing countries and is unattractive in poor countries [10].

A significant contribution to atmospheric pollution is made by heat and power stations (hereinafter referred to as CHP) operating on traditional fuel sources (coal, oil, and gas); their share (Figure 1) is about 60% of the global electricity generation [11]. The issue of using renewable energy sources (RES) for electricity generation is relevant, considering the trend programs of many developed countries towards the reduction of carbon dioxide emissions into the atmosphere and providing access to inexpensive, reliable, sustainable, and modern energy for all segments of the population [12]. These include solar panels, wind generators, and biofuels [13].

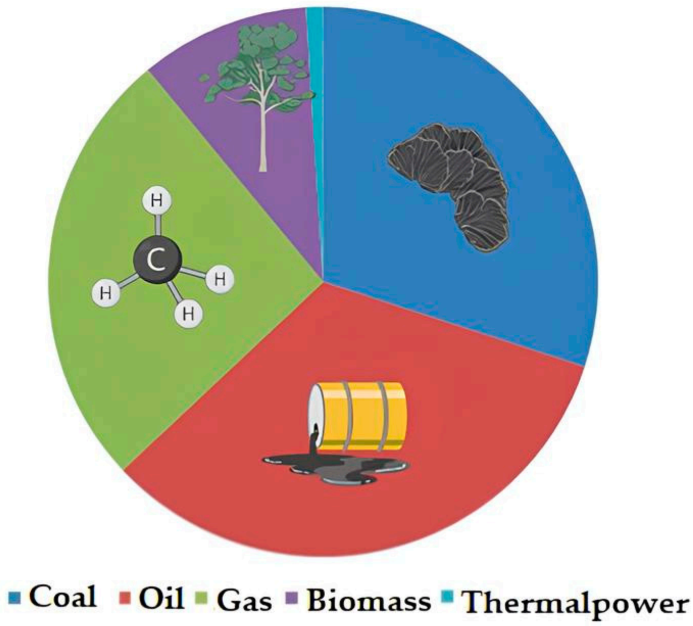

Figure 1. Global electricity generation by type of fossil fuel (according [14]).

However, renewable energy sources have a number of disadvantages. Thus, the process of their disposal is an extremely difficult task [15] since, for example, solar panels contain elements such as As, Cd, Hg, and Pb [16], which can have a negative impact on the ecosystem, and their burial is an extremely undesirable method of their disposal [17]; thermal and chemical methods of solar panel recycling have not been sufficiently mastered and are not characterized by a high degree of efficiency [18,19]. Dust is released during the mechanical processing of solar panels. It contains glass fiber, noise pollution is created, and rare earth elements are lost. However, 80 million tons of waste from used solar panels are expected worldwide by 2050, which will inevitably have a negative impact on the surrounding ecosystem.

The use of biofuel cells (BFCs) is an effective alternative in this context, as electricity generation is carried out in the process of the biocatalytic oxidation of various substrates. Despite the low power generated in the BFC system and, as a result, a long payback period, the research in the field of biofuel elements is relevant due to humanity's awareness of global environmental problems, the need to solve which reduces the role of economic levers in the development of the world community. PubMed (NCBI) has pointed to an exponential growth of publications on the subject of a "biofuel cell" in the first decade of our century, and this interest persisted throughout the following years. It should be noted that biofuel elements based on microorganisms (microbial fuel cells, MFCs) are a promising technology to produce bioelectricity since they simultaneously solve the problems of contamination with anthropogenic organic waste, which can be used by microorganisms as a source of carbon and energy. A continuous and steady supply of organic substrates is required to ensure the uninterrupted generation of electricity in an MFC, which cannot always be implemented in practice. A fairly new technology of plant microbial fuel cells (hereinafter referred to as PMFCs) eliminates this disadvantage of MFCs largely. The electricity generation is carried out via the oxidation of organic substances using microorganisms that are both synthesized in plants during photosynthesis under the action of sunlight energy and produced into the environment (root exudates, root deposits, and rhizo-deposition) and come from outside, for example, from wastewater or industrial waste. Such hybrid energy technology can be used in phytomonitoring the state of plant crops, a local power supply, charging portable devices [20], powering various low-power sensors to monitor ambient temperatures and humidity, power camera traps in remote areas [21], and serve as a biosensor for monitoring plant health in smart greenhouses [22] (Figure 2). It should be noted that the PMFC technology, using macrophytes, reduces the level of greenhouse gases (N_2O and CH_4) by 5.9–32.4% in terms of CO_2 [23].

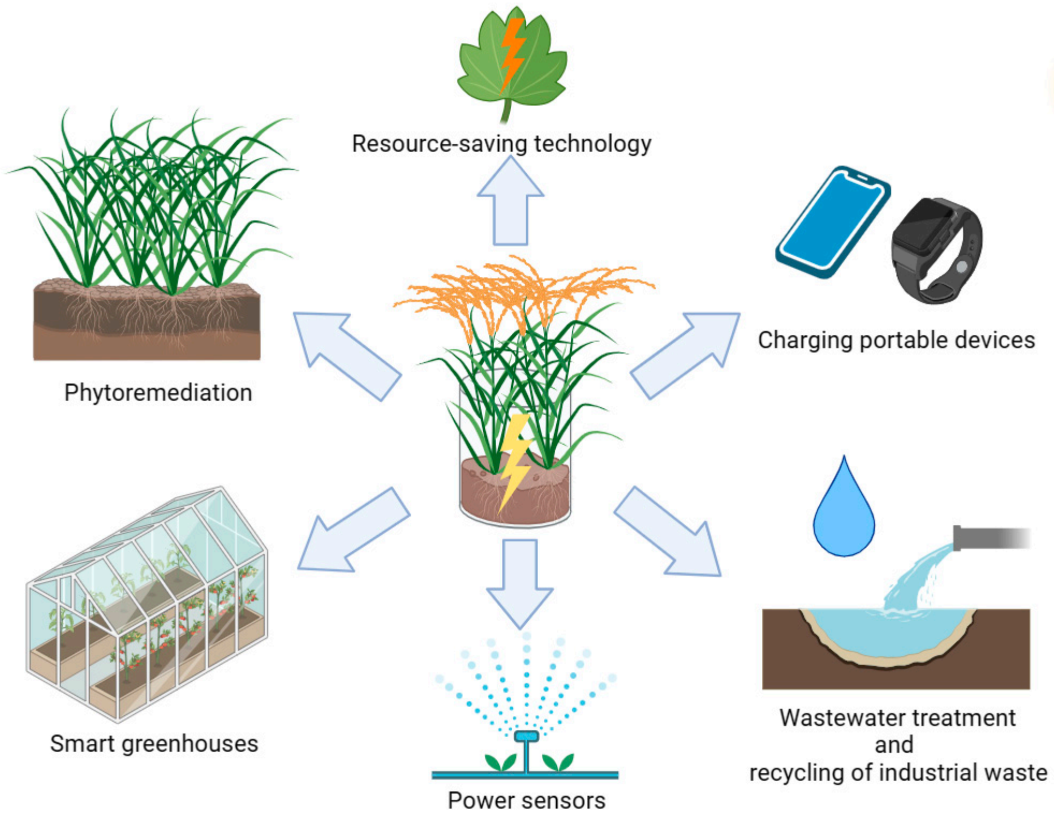

Figure 2. Possibilities of PMFC application.

The review [24] summarizes the evolution of PMFC technology and discusses the basic principles associated with it, factors affecting its effectiveness, application areas, prospects, and disadvantages. The review [25] describes in detail such features of a PMFC as its basis, the function of plants and their rhyzo-deposition, electrical characteristics, internal resistance, substrate kinetics, redox reactions of the root medium, and electron transport mechanisms. This work [25] also depicts the used microbial communities capable of electrogenesis, and it presents the most common PMFC structures and a comparative analysis of their characteristics. The issues of PMFC technology usage for wireless energy sensing, farming, and agricultural applications of the next generation are highlighted at the end of the review. However, the existing reviews pay little attention to PMFC technology usage to solve the environmental problem of anthropogenic pollutant utilization from water and soil ecosystems along with electricity generation.

This review briefly describes the principle of the PMFC technology's functioning and the influence of environmental factors on the PMFC characteristics; our emphasis is on the environmental aspect of the PMFC technology's application.

2. Plant Microbial Fuel Cells: Functioning and Factors Affecting the Electrochemical Characteristics of the SYSTEM

The generation of electricity depends on many factors, such as the types of exo-electrogenic microorganisms used, the material of the electrodes and their modification, environmental factors, and the plants used. Understanding the functioning principles and the optimal choice of microorganisms and plants makes it possible to increase the efficiency of electricity generation in a PMFC.

2.1. The Principle of PMFC Operation

The principle of PMFC operation is based on two interrelated processes: the synthesis of rhizo-deposits in plants and their use as a substrate by microorganisms to generate electricity (Figure 3). Complex interactions in heterogeneous, polydisperse, multifactorial natural systems were previously described as a computer model of the chemical and microbiological production processes of plant biomass, soil microorganisms, and nutrients in the rhizosphere [26].

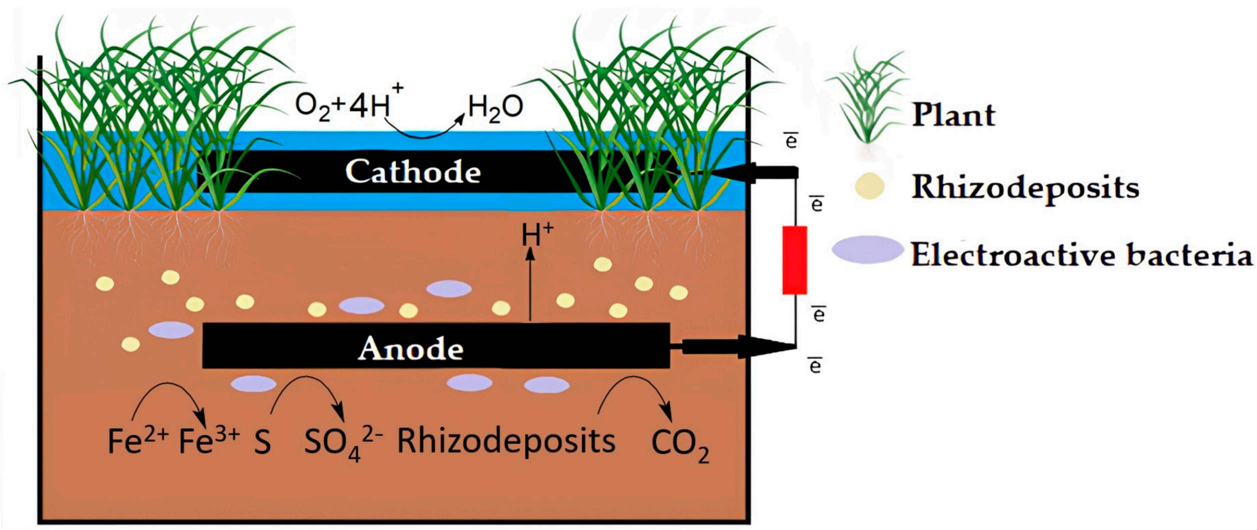

Figure 3. The scheme of PMFC functioning.

Photosynthesis regulates the vital activity of plants, during which plants fix carbon dioxide from the atmosphere and form carbohydrates, organic acids and amino acids, secrets—polysaccharide mucus (mucigel), lysates—materials of dead cells, gases—ethylene ethylene and carbon dioxide under the influence of sunlight energy [27]. Electrogenic microorganisms use deposits as substrates for growth and development, as well as electricity generation as a result of ongoing oxidative processes involving the enzymatic systems of microorganisms. As a result, carbon dioxide is synthesized, and free charge carriers (protons and electrons) are formed. Charges need to be separated to convert chemical energy into electrical energy. The process is carried out by moving the generated electrons at the anode to the cathode through an external circuit; protons migrate through a nutrient matrix or medium from the substrate to the cathode due to the presence of a potential gradient [18], where molecular oxygen or another catalyst and water molecules are formed [28]. However, it is likely that hydroperoxyl radicals (HO_2) are formed on the cathode during the reduction process as an intermediate product [29]. Microorganisms, in turn, can enter symbiosis with plant roots, forming protective biofilms and producing antibiotics to protect plants from pathogens [30].

When choosing microorganisms, it is necessary to consider their ability to transfer electrons to the anode (Figure 4), which can be caused by various mechanisms: direct electron transfer through cytochromes and electron-conducting molecular saws (nanowires) with the help of electroactive compounds (mediator transfer). General information about this various mechanisms is summarized in recent reviews and articles [31–37].

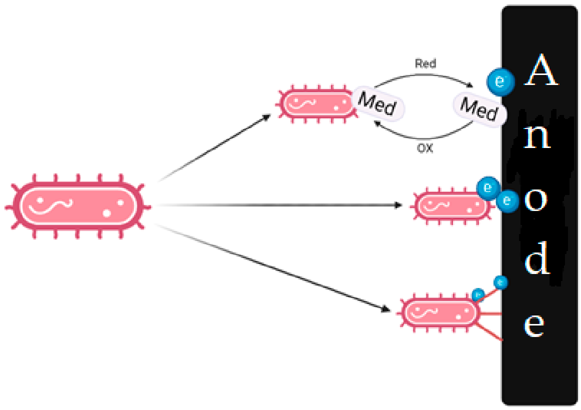

Figure 4. Various ways in which electron transfer to the anode can occur.

The focus is on natural ecosystems when choosing microorganisms for a PMFC system. Relatedly, bacteria inhabit the environment in the rhizosphere; they are anaerobes that produce protons and carbon dioxide and can transfer electrons to the anode during the oxidation of organic compounds. Table 1 presents a description of some rhizospheric bacteria.

Table 1. Rhizospheric microorganisms capable of direct extracellular electron transfer.

Microorganism	Description	Consumable Substrates	References
Desulfobulbus sp.	Obligate anaerobes capable of oxidizing sulfur to sulfate using an anode as an electron acceptor.	Acetate, propionate, butyrate, lactate, and pyruvate	[38–40]
Geobacter sp.	Anaerobic metal-reducing bacteria. Fe (III) and Mn (IV) are used as electron acceptors. They can transmit electrons using pili—filamentous protein formations.	Benzoate, p-cresol, trichloroethane, benzene, lactate, acetate, and starch	[41]
Geothrix fermentans	Anaerobic metal reducers. Fe (III) is used as an electron acceptor. They are capable of forming extracellular mediators of the quinone series and riboflavin, which makes it possible to transfer electrons to the electrode more efficiently.	Acetate, propionate, lactate, and fumarate	[42,43]
Rhodoferax ferrireducens	Facultative metal-reducing anaerobe with a wide temperature range of growth. Fe (III), Mn (IV), nitrate, fumarate, and oxygen can be used as electron acceptors.	Acetate, lactate, propionate, pyruvate, malate, succinate, and benzoate	[44]
Shewanella sp.	Facultative anaerobic bacteria using Fe (III) and Mp (IV) as electron acceptors are capable of producing flavins that act as electronic transfer mediators.	Lactate and formate	[45,46]
Clostridium butyricum *C. beijerinckii*	Obligate anaerobes can use an anode as an electron acceptor. Hydrogen, which is able to oxidize at the anode, is produced during the enzymatic fermentation of substrates.	Glucose, starch, sucrose, and lactate	[47]

The basic property of microorganisms that allows their use in bioelectric systems [48–50] is their ability to produce electroactive compounds, as well as to use an anode as an electron acceptor. Moreover, the use of inorganic anions as an electron acceptor makes it possible to reduce the salinity of treated wastewater [51,52], for example, when using sulfate-reducing bacteria that are capable of the assimilatory reduction of sulfates to sulfides [53].

PGPR (plant-growth-promoting rhizobacteria), which promote plant growth, play an important role in maintaining the vital activity of plants and are used for the development of PMFC. Such microorganisms include, for example, bacteria of the species *Bacillus thuringiensis*, which are involved in nitrogen fixation processes, sulfur and phosphorus exchanges, and the synthesis of plant growth stimulants [54]. Bacteria of the genus *Pseudomonas* sp. can be also considered as a PGPR-group bacteria [55]. Some species of *Pseudomonas* sp. are capable of surfactant destruction [56,57]; they can form biofilms on the surface of an anode and secrete compounds of the phenase-new series [58]. These compounds play an important role both in protecting plants from pathogen infection [59] and stimulating the growth of shoots [60]. Moreover, phenazines act as mediators of the electronic transport between bacteria and an electrode [61]. Bacteria of the family *Ruminococcaceae* spp. are not electroactive but are capable of utilizing cellulose (35–50% of the dry plant weight) while producing organic substrates, which are additionally used by electroactive microorganisms as electron donors [62]. Therefore, the use of PGPR-group bacteria can be used in PMFC systems to stimulate plant growth and protection, which theoretically can have a beneficial effect on electricity generation.

2.2. Electrodes in PMFC

It is important to choose the right electrode material for the efficient generation of electrical energy when creating PMFCs along with biological components [63]. Generally, the electrode material should have high electrical conductivity, electrochemical stability, porosity, and biocompatibility [64]. Metals (zinc [65], stainless steel [66], and platinum [67]) and carbon materials [68] are usually used as electrodes in bioelectrochemical systems. Despite the high electrical conductivity of metals in comparison with carbon materials, the use of stainless steel, for example, increases the period of microorganism adaptation on the metal anode surface [68]. It causes a decrease in current generation at the initial stage of the PMFC operation, which is explained by the lower biocompatibility of stainless steel to microorganisms. Moreover, metals are subject to corrosion processes [66] and have a high cost, thus limiting their use in PMFC development.

The geometric area of the electrodes affects the output of electricity—the larger the area, the more contact there is for electroactive microorganisms, which leads to an increase in current density [69]. In turn, graphite electrodes (felt/fiber) have a developed surface that promotes the adhesion of microorganisms and the sorption of organic compounds. This material is not subject to corrosion; therefore, it is promising for the creation of PMFCs [70]. The addition of granular graphite or activated carbon to the surface of the anode improves the adsorption of organic compounds and increases the specific surface area for colonization via bacteria. Electrode modification is used to improve the producible power of bioelectrochemical systems, which is described in detail in recent articles [71–75]. The use of carbon materials produced from crop waste is also promising in this field [76].

Thus, the choice of electrode material is the key element determining the efficiency of the entire PMFC system. Existing materials can be modified to reduce their internal resistance in order to increase the current output and power.

2.3. Application of Proton Exchange Membranes in PMFC System

Various PMFC configurations have been developed so far: sediment PMFCs, constructed-wetland MFCs, tubular PMFCs, floating-treatment wetland MFCs, flat plate PMFCs, and power-generating trees. The advantages and disadvantages of each model are detailed in the review [24]. One of the components of bioelectrochemical systems for power generation is a proton exchange membrane, which allows the improvement of charge segregation and power performance [77]. The most preferred proton exchange membrane is Nafion, but its use in BES significantly (by 40%) increases the cost of the device [78]. Thus, the search for new membranes that will have a lower cost and provide high stability and efficiency in BES is currently underway.

In [79], modified Nafion 117 proton exchange membranes were tested. The modification included the treatment of the membrane with solutions of polyvinylidene difluoride (PVDF) and sulfonated PVDF with the addition of silicon oxide (SiO_2). The third modification involved the polymerization of a Nafion membrane in a methyl methacrylate (MMA) solution with the addition of sodium sulfite as an initiator. According to the results obtained, all three methods increase the power generation parameters of MFC systems. The highest increase in current density, from 0.81 mA/m^2 to 18.82 mA/m^2, was demonstrated using the modification of Nafion with MMA.

In [80], a proton exchange membrane based on agar and polyvinyl alcohol (PVA) with the addition of vermiculite nanoparticles was tested. According to the results obtained, the proton exchange properties of the tested membranes were 216% higher than those of the commercial Nafion membrane. In addition, the MFC current density increased (from 605 mA/m^2 to 1515 mA/m^2) when agar and PVA-based membranes were used. A low cost and environmental safety, in combination with the increased efficiency of MFC energy generation, allow the use of agar and PVA-based membranes as an alternative to expensive Nafion membranes.

Ceramic membranes based on clay, bentonite, coal ash, Na_2CO_3, Na_2SiO_3, and H_3BO_3 were considered in [81]. The use of hybrid ceramic membranes with the addition of different compounds contributed to the increase in PMFC power density by 78% (up to 22.38 mW/m^2) compared to the control (100% clay membrane). There was a decrease in internal resistance from 346 Ω (control) to 234 Ω. The addition of bentonite, coal ash, Na_2CO_3, Na_2SiO_3, and H_3BO_3 improved the membrane's cation transport, reducing oxygen diffusion to the anode chamber. The membrane demonstrated high stability during 6 months of PMFC operation. In addition, the ceramic membrane is significantly cheaper than the Nafion membrane.

Thus, one of the important aspects of PMFC operation, power increase, and internal resistance reduction is the use of proton exchange membranes. At the same time, for the commercialization of PMFC systems, it is necessary to take into account the cost of the production of such membranes and the expenses associated with the complication of the design when using membranes.

2.4. The Influence of Environmental Factors on the Electricity Generation in a PMFC

The metabolic activity of exoelectrogenic microorganisms, which play an important role in BES functioning and electricity generation, depends on the temperature, the pH, and the rate of organic substrates' receipt. Thus, the work [82] showed that, when the air temperature rises to 30 °C, the voltage of the bioelectrochemical system increases from 100 to 150 mV, which may be due to an increase in the metabolic rate of exoelectrogenic microorganisms. The pH value affects the development of microorganisms. pH of 6–9 is mostly suitable for the functioning of BES [83]. The power decreases to 158 mW/m^2 at a pH value of 6.0 for the MFC system [84], while the power value is 600 mW/m^2 at a pH of 8.0. The inhibition of the metabolic activity of exoelectrogenic microorganisms is observed with a decrease in pH, which contributes to a decrease in the BES power [85].

Periodic watering is necessary for the normal functioning of plants since soil moisture affects the generated potential in a PMFC system. The article [86] states that, in the absence of irrigation, the soil dries up, which leads to a two-fold decrease in the PMFC potential, but after watering (60–70% of the soil moisture capacity), the potential is restored. Thus, energy generation changes depending on the time of day [87]. An increase in electrogenic activity is observed after sunrise due to the launch of photosynthesis processes, the peak of which is observed from 14 to 15 h. Depending on the system under study, the open circuit potential is 600–700 mV at the specified time. Then, the photosynthetic activity of plants decreases at nightfall, which leads to a decrease in electricity generation to 300–400 mV.

The rate of photosynthesis is affected by the concentration of carbon dioxide in the atmosphere [88]. The trend towards carbon dioxide emissions increases every year and is 390 ppmv, according to the latest data (mass fractions of a percent per volume), which is 30% more than the CO_2 concentration in the early twentieth century [89]. The increasing CO_2 concentration and climate warming significantly affect plant growth [90]. The work [91], using agricultural plants (*Saccharum officinarum* and *Sorghum bicolor*), showed that the rate of photosynthesis grows significantly with an increase in the CO_2 concentration, which in theory can have a positive effect on the power produced via a PMFC. It should be noted that plants with the C_3 and C_4 types of photosynthesis react differently to an increase in the carbon–acid gas concentration. C_4 plants attach CO_2 to phosphoenolpyruvate [86], resulting in the formation of oxalic acid containing four carbon atoms. The photosynthesis efficiency of C_4 plants is significantly higher since the C_4 pathway is an extra pump that supplies additional portions of CO_2, increasing its concentration in the plant since the CO_2 concentration in the assimilation chamber is lower than in the air, which is a limiting factor of photosynthesis.

It should be noted that the countries with warm climates and high solar insolation, as well as "green roofs" cities, have the greatest potential for the PMFC technology's implementation to reduce the concentration of carbon dioxide in the air [92].

3. PMFC Technology to Utilize Anthropogenic Pollutants in Aquatic and Soil Ecosystems: Current Situation and Further Development

3.1. PMFC to Purify Water and Soil Ecosystems from Organic Compounds and Biogenic Elements

Wastewater discharges, containing organic and biogenic (nitrogen, phosphorus, and carbon) elements in concentrations above the MRL (maximum residue limit), significantly affect the ecological balance of aquatic ecosystems (Figure 5). Thus begin the eutrophication and rapid development of microbiota, which entail a decrease in the dissolved oxygen concentration, causing a decrease in biological diversity [93]. Additionally, the current active use of oil has a negative impact on ecosystems that have been polluted by its spills during production and transportation. The sludge formed during oil production is discharged into specialized ponds, which "age" under the influence of the environment. So, the oxidation of some components, tarring, and the evaporation of light fractions occur. These processes lead to increased stability of the oil sludge in purification; therefore, its disposal is one of the most difficult tasks at present [94].

Bioremediation, based on pollutant biodegradation via microorganisms and plants during their vital activity, is one of the most effective methods of wastewater and soil treatment for organic pollutants. It is used for wastewater treatment and the processing of biodegradable solid household waste to form biogas [95,96]. As was noted, PMFC technology is promising in the simultaneous processes of generating electricity [21] and recycling various pollutants [97]. Table 2 summarizes the information on the developed PMFC for the disposal of anthropogenic pollutants and their purification.

The authors of [98] used *Pseudomonas*, *Azoarcus communis* oil destructor bacteria to clean the soil from oil pollution. The addition of bacteria to the PMFC system contributed to the better purification of wastewater and soils from hydrocarbons. The maximum specific power (11.56 mW/m^2) was obtained in a system where *Spartina sp* was used as a plant. It was almost five times higher compared to the control system without plants, the value of which reached about 2 mW/m^2. Generally, power increases (7.5 mW/m^2 and 9.71 mW/m^2, respectively) appeared because of the use of *Typha latifolia* (broadleaf cattail) and *Phragmites* (common reed) plants due to the formation of rhizodeposits that could be consumed by microorganisms. The internal resistance of the studied systems ranged from 200 Ω to 400 Ω.

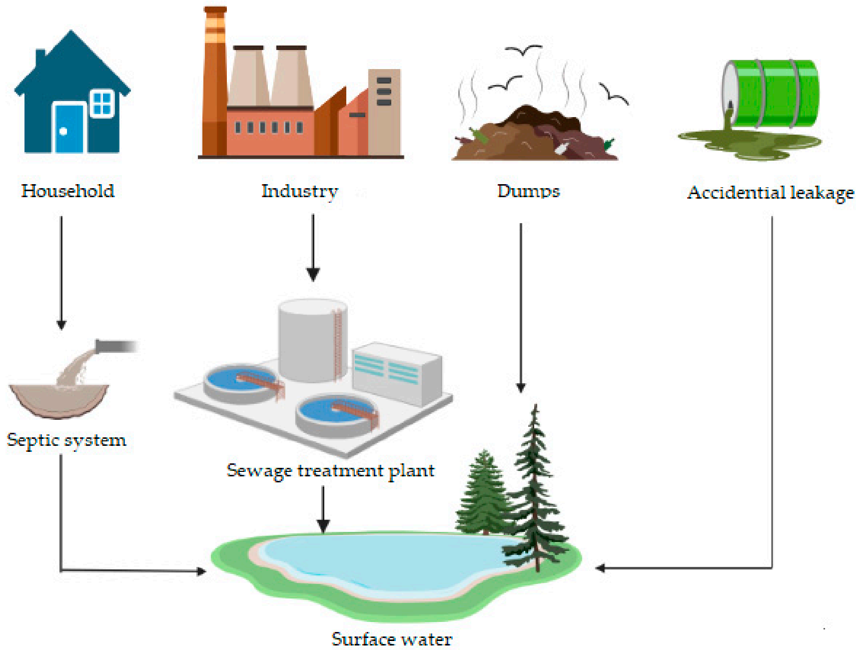

Figure 5. Inflow of anthropogenic pollutants into aquatic ecosystems.

Table 2. PMFC characteristics for soil and wastewater treatment with organic compounds.

Plants	Microorganisms	Electrode Material	Organic Compound/ Rate	Purification Rate, %	Max. Output	Ref.
Spartina sp.	*Pseudomonas veronii* *Ps. chlororaphis* *Ps. putida* *Ps. libanensis* *Azoarcus communis*	Cathode—stainless steel Anode—stainless steel	Oil	99.6	11.56 mW/m^2	[98]
Aglaonema commutatum	Active sludge	Cathode—carbon felt Anode—carbon felt	Oil PAC	Up to 82.3 Up to 45.5	382 mV 377 mV	[99]
Steviare baudiana	Soil extraction	Cathode—stainless steel Anode—carbon felt	Urea	No data	132 mW/m^2	[100]
Ozyra sp.	Soil extraction	Cathode—carbon felt Anode—carbon felt	Compost	No data	39.2 mW/m^2	[101]
Fimbristylis ferruginea	Association DC5 (*Firmicutes* *Proteobacteria* *Bacteroidota* *Desulfobacterota* *Actinibacteriota* *Verrucomicrobiot)* Soil extraction	Cathode—glassy carbon fiber Anode—glassy carbon fiber	Textile wastewater	Up to 97.3	Up to 197.9 mW/m^2	[102]
Canna generalis, Chrysopogon zizanioides, Cyperus papyrus Hymenachne grumosa Equisetum hyemale	Wastewater bacteria	Cathode—graphite Anode—graphite	BOD_5 COD	71 74	0.93 mW/m^2	[103]
Chlorella vulgaris	Anaerobic sludge	Cathode—carbon felt Anode—carbon felt	COD Nitrates Phosphates	65.3 66.6 95.6	3.64 mW/m^2	[104]
Canna indica		Cathode—carbon felt A—carbon felt	COD Nitrates Phosphates	57.2 59.8 88.8	22.76 mW/m^2	
Schoenoplectus californicus	Sludge	Cathode—activated carbon Anode—activated carbon	COD Nitrogen	Up to 87 Up to 98	8.6 mW/m^2	[105]
Canna indica	Anaerobic sludge	Cathode—stainless steel, activated carbon Anode—stainless steel, activated carbon	Tetracycline Sulfatotoxal	99.66 100	Up to 124.89 mW/m^2	[106]
Canna indica	Soil extraction	Cathode—graphite plate Anode—graphite rod	Sodium dodecyl benzene sulfonate	Up to 56.8%	4.01 mW/m^2	[107]

The low solubility of petroleum products is one of the problems of effective purification from them. Surface-active substances (surfactants) are additionally introduced into PMFC systems to increase the bioavailability of the hydrophobic substrate for oil-oxidizing bacteria and, therefore, to increase the degree of biodegradation of oil pollution. It allows an increase in the voltage from 184.9 to 377.2 mV [99]. Moreover, the introduction of an additional carbon source available to microorganisms, for example, glucose, contributes to an increase in the system voltage from 184.9 mV to 325 mV [98]. Biosurfactants should be used instead of using synthetic surfactants (surfactants). Biosurfactants can increase the bioavailability of hydrocarbon substrates to change the hydrophobic properties and permeability of microorganisms' membranes [108]. Moreover, oil-resistant plants should be used to clean the soil from oil in PMFC systems. They can release a high amount of root exudates. It follows that further directions in the development of new soil and wastewater treatment systems may consist in the selection of effective bacteria–oil destructors.

PMFC systems can be used for the disposal of organic animal waste. The work [100] used the plant stevia honey and urine samples of livestock (goats, cows, and sheep). It was noted that the addition of urine stimulated plant growth and significantly increased the current density of the device. The control sample had fixed the current density at about 10 mA/m^2, and this indicator reached a value of 930 mA/m^2 with the addition of cow urine. Thus, it was noted that the use of cattle urine is a good tool aimed at increasing PMFCs' characteristics.

Wastewater treatment uses the technology of microbial associations' enrichment with bacteria that are isolated from contaminated soils. For example, the authors of [102] used an association of microorganisms, designating it as DC5. These microorganisms are capable of oxidizing textile dyes, which can potentially act as a mediator of electronic transport, thus contributing to an increase in electricity generation during their biodegradation [109]. The addition of the DC5 association to the PMFC contributes to an increase in the maximum specific power from 177.3 mW/m^2 to 197.94 mW/m^2. This approach makes it possible to improve wastewater treatment from electroactive dyes and at the same time to increase the electricity generation in PMFC systems.

A plant microbial fuel cell is a promising system for wastewater treatment. Thus, the work [103] used a reactor for anaerobic purification where wastewater enters the PMFC. A significant decrease in suspended compounds was observed after two-stage purification. The treated wastewater, taken from the PMFC, had a BOD_5 index lower than the initial one by 71%; the COD index decreased by 74%.

The plants used can influence the efficiency of purification and the generation of electricity in a PMFC. The review [104] studied the effectiveness of wastewater treatment using the *Canna indica* plant (Indian cane) and single-celled algae *Chlorella vulgaris*. The system based on the *C. indica* plant had a higher voltage (771 mV and 452 mV, respectively). The internal resistance for a system with a plant was about 100 Ω, with chlorella algae at 335 Ω. At the same time, the paper notes that *C. vulgaris* can decompose organic compounds, indicating a mix-trophic type of nutrition that provides better purification from organic compounds. Conversely, *C. indica* has an autotrophic type of power supply, and the degradation of organic compounds occurs at the anode when electroactive microorganisms are introduced into the system, contributing to a higher voltage output and specific power of the system.

The work [106] investigated the possibility of removing two antibiotics, tetracecline and sulfamexosol, from wastewater in a PMFC system. According to the obtained data, the greatest degree of removal was achieved during the first day of the PMFC operation. The removal efficiency reached 99%. It was shown that both antibiotics can accumulate in insignificant amounts in electrode compartments, which is due to electrosorption [110]. It occurs because of the formation of a double electric layer (DEL) on the surface of cathodes and anodes, while tetracecline and sulfamexosol do not accumulate in plants. In systems with *C. indicia*, the specific power is, on average, 55% higher than in systems without plants. The resulting power density is 124.89 mW/m^2, and the internal resistance ranges from

600 Ω to 800 Ω for all systems. Generally, the conducted research testifies to the prospects of PMFC systems in the removal of pharmaceutical preparations from wastewater.

A *Canna indica* plant-based PMFC system was described in [107], which was used to remove sodium dodecyl beznesulfonate (SDBS) from a model wastewater mixture. According to the results, the removal efficiency of SDBS was 56.8%, and power values of 4.01 mW/m^2 and voltage of 230 mV with a resistance of about 200 Ω were achieved at an SDBS concentration of 5 mg/L. It should be noted that increasing the SDBS concentration had a negative effect on the PMFC systems, reducing the power and increasing the internal resistance.

It should also be noted that, in these works, practically no attention was paid to the problem of wastewater treatment in a PMFC for surfactants. However, the negative impact of surfactants on the environment was ignored because of the large scale of production and use [111]. Therefore, currently, there are a great number of studies focusing on the ways surfactant disposal can be achieved in wastewater, including the development of approaches to decentralized treatment methods [112,113]. Generally, surfactants have a negative effect on aquatic biota at concentrations ranging from 0.35 mg/dm^3 (freshwater microalgae *S. subspicatus*) [114] to 76.14 mg/dm^3 (crustaceans *D. magna*) [115]. The entry of surfactants with wastewater into natural reservoirs can have a negative impact on the cultivation of aquaculture in natural conditions. For example, the review [116] investigated the influence of anionic and cationic surfactants on the survival, behavioral features, and viscera pathology of *African catfish Clarias gariepinus*, used as a test object. Anionic surfactants had the greatest negative effect (linear alkylbeznosulfonate (LAS) was used as a model toxicant). The behavioral characteristics of the test object included excessive mucus secretion, chaotic movement, and anger. The lethal concentration of LAS equaled 10.57 mg/dm^3. Gill injuries were caused by the constant contact of this organ with the environment and surfactants [117,118]. Biological wastewater treatment from surfactants, when activated sludge is used, provides 95–98% efficiency [119,120]. Microorganisms use surfactants as a carbon source, while biodegradation occurs along the pathways of ω-oxidation, β-oxidation, α-oxidation, and oxidation of the benzene ring (if it is presented in surfactants) [121]. The bacteria that can oxidize surfactants include *Azotobacter* sp., *Bacillus* sp., Pseudomonas sp., *Citrobacter* sp., *Acinetobacter* sp., *Klebsiella* sp., and *Serratia* sp. [122–126]. Therefore, the selection of microorganisms that can simultaneously reduce the concentration of surfactants and achieve exoelectrogenicity is an urgent task in PMFC development to purify wastewater from surfactants.

Several works on MFC have demonstrated the combination of wastewater treatment from surfactants and electricity generation [127–129]. The presented works demonstrate that water purification in bioelectrochemical systems from surfactants at concentrations from 10 to 120 mg/dm^3 of compounds is possible. The efficiency of this process can reach up to 90% and depends on the time and structure of the surfactant. In general, the process of surfactant biodegradation takes from 12 to 96 h. However, high internal resistance negatively affects the power output of BES [130].

Thus, PMFC can be used for the treatment of wastewater that may contain not only biogenic elements but also antibiotics and petroleum hydrocarbons, including PAHs. At the same time, a note should be made concerning the selection of the optimal composition of the microorganism association to reduce the time of their adaptation to pollutants and higher electricity generation. It bears emphasis that there are fluctuations in the internal resistance of various systems (from 100 Ω to 800 Ω), which are associated with different designs, the distance between the anode and cathode, and the electrical conductivity of the electrolytes used. Nevertheless, it should be noted that there are rather few works that have dealt with the treatment of wastewater from surfactants using PMFC systems.

3.2. PMFC Application for Removal of Heavy Metals from Soil and Aquatic Ecosystems

Soil pollution with heavy metals (HM) poses a threat to the environment and agriculture [131]. Heavy metals negatively affect agricultural crops, reducing their yield.

Phytoremediation methods are used to clean soils of heavy metals, the principle of which is based on biosorption and HM accumulation by various plant components. The removal rate of heavy metals from soils is about 35% of the initial HM concentration during the soil phytoremediation [132]. Microorganisms are also able to reduce concentrations of HM ions by forming chelated complex compounds with them, which is due to the production of siderophores, organic acids, and extracellular polymeric substances [133]. However, as has been shown earlier, bioremediation has not become widespread in poor countries due to its relatively high cost [10]. Therefore, the use of PMFC technology can become a compromise solution in poor countries not only because of the purification of contaminated soils from HM but also due to the generation of environmentally friendly electricity [134]. Table 3 presents the parameters of some well-known PMFC systems used for soil purification from heavy metals.

Table 3. PMFC used for soil purification from heavy metals.

Plant	Microorganism	Electrode Material	Metal	Purification Rate, %	Max. Generation	Ref.
Lolium perenne	*Proteobacteria* *Bacteroidetes* *Firmicutes*	Anode—graphite granules and carbon felt Cathode—carbon felt	$Cr_2O_7^{2-}$	90–99	55 mA/m^2	[135]
Oryza sativa L.	*Alphaproteobacteria* *Anaerolineae* *Clostridia* *Deltaproteobacteria* *Gammaproteobacteria* *Actinobacteria* *Bacteroidia* *Bacilli*	Anode—carbon felt Cathode—carbon felt	As (V)	25.2–41.8	22.2 mW/m^2	[136]
Oryza rufipogon *Typha orientalis*	*Thermoleophilia* *Nocardioides* *Anaerolinea* *Geobacter* *Tumebacillus* *Azospirillum* *Bacillus*	Anode—carbon felt Cathode—carbon felt	Cd (II)	Up to 31.7 Up to 30.2	351 mV 137 mV	[137]
Eichhornia crassipes	No data	Anode—graphite rod Cathode—graphite rod	Ni (II)	Up to ~10	0.86 mW/m^2	[138]
Oryza sativa L.	*Proteobacteria* *Firmicutes* *Actinobacteria* *Chroroflexi*	Anode—carbon felt Cathode—carbon felt	Cd Cu Cr Ni	35.1 32.8 56.9 21.3	22.2 mW/m^2	[139]
Cyperus alternifolius *Cyperu smalaccensis*	River sludge	Anode—carbon felt Cathode—carbon felt	As Zn Cd	6.7 7.3 38.5	10.74 mW/m^2	[140]

Raygrass has been used to remove $Cr_2O_7^{2-}$ from a PMFC system [135]. According to the results, the removal efficiency can reach 99% under various conditions. At the same time, most of the precipitates reduced to Cr (III) took the form of $Cr(OH)_3$. Meanwhile, an increase in the concentration of $Cr_2O_7^{2-}$ from 9 mg/dm^3 to 19 mg/dm^3 increased the current density by about two times (up to 55 mA/m^2).

The study [136] tried to reduce the absorption of soap using the rice culture *Ozyra sativa L.* (seeded rice) since rice consumption is one of the main routes of arsenic's entry into the human body. The results show that the use of PMFC technology reduces the arsenic accumulation in rice by up to 67.9% due to the obstruction of As (III) migration to the plant roots. The output power equals 22.2 mW/m^2.

The article [137] illustrates the PMFC development based on *Ozyra rufipogon* (wild rice) and *Typha orientalis* (oriental cattail) to remove cadmium from the soil. It was shown that cadmium absorption is carried out mainly via plant roots. The addition of biochar contributed to the better removal of cadmium from the soil. The use of PMFC reduces cadmium mobility by binding to carbonates, iron oxides, and organic compounds. When a PMFC operates in a closed-circuit mode, it was noted that the percentage ratio of the Cd^{2+} exchange fraction is significantly lower than when the PMFC operates in the open circuit mode. This is caused by bioelectrochemical processes. Additionally, higher values of the generated voltage were observed (350 mV vs. 137 mV) when using rice. At the same time, the voltage decreased from 400 mV to 150 mV in the control system (without Cd (II)) with cattail by 112, and the voltage increased to 400 mV in the system with rice by the end of the experiment. Such a feature may be due to the different compositions of the rhizodeposits.

The study [139] used seeded rice exploitation with HM such as, Cd, Cr^{-}, Cu, and Ni. It shows that a decrease in the concentration of HM in PMFC (compared with the control rate) was possible to be achieved during the experiment. The authors noted that the removal of such heavy metals as Cu was probably due to the transition under the action of an electric field of Cu to the cathode region, where they react with oxygen and precipitate in the oxide form. Besides, bacterial biofilms can absorb Cu through the cell membrane. Similar mechanisms of reduced mobility were observed for Cr and Ni. The maximum current is 1.20 mA, while the fluctuations of this value were caused by a change in the oxygen volume in the anode compartment from the rice roots. The generated capacity of 22.2 MW/m^2 allowed the conclusion that the PMFC system can be applied to soil purification from some heavy metals in their joint presence.

The above data prove that soil purification from heavy metals in PMFC systems is practically not inferior to phytoremediation. In some cases, a decrease in the mobility of some HM [140] can be detected because of their conversion into poorly soluble compounds due to the course of bioelectrochemical processes in PMFC systems.

Thus, the use of PMFC technology to combat soil pollution still needs further study since the processes and mechanisms that occur should be considered separately for various heavy metals and plants. The use of PMFC, according to the research, is promising due to the economic effect that is caused by electricity generation and low-cost phytoremediation technologies [141].

3.3. Comparison of Treatment Efficiency of PMFC Technology with Traditional Methods

This section provides estimates of the effectiveness of the PMFC systems described above compared to conventional approaches to wastewater treatment. Table 4 summarizes the physicochemical, physical, and biological methods.

Table 4. Comparative analysis of conventional wastewater treatment technologies and PMFC systems.

Treatment Method	Comments	Monitored Indicator	Treatment Efficiency, %		Ref.
			Current Method	PMFC	
Physicochemical	Electrooxidation and electrocoagulation	COD	90	57–87	[142]
		Nitrate	97	59–67	
		Phosphates	90	88–95	
Physical	Membrane filtration	COD	60	57–87	[143]
		BOD	65	71	
	Adsorption	Cr (VI)	84	57–99	[144]
Biological	Microalgae *Chlorella vulgaris*	COD	100	57–87	[145]
		BOD	96	71	
		Total nitrogen	61	89	
	Biofilter with immobilized bacteria and macroalgae	COD	69	57–87	[146]
		Total nitrogen	59	89	
	Active sludge	SDS	100	57	[147]
		COD	up to 91	57–87	

In [142], electrocoagulation and electrooxidation using electrodes made of Al and Fe in combination with diamond, which was doped with boron, were used for wastewater treatment. The high efficiency of these physicochemical methods of wastewater treatment for such indicators as COD, phosphate, and nitrate concentration should be noted. Compared to the above-described PMFC systems, the method of the electrooxidation and electrocoagulation of nitrate is faster (treatment time—60 min).

In [143], a physical method of water purification was used, employing a membrane made of polyethersulfone and nanocrystalline cellulose (NCC), which was fabricated via phase inversion. Three membrane samples with different cellulose contents (0%, 1%, and 5%) were prepared. According to the results, the introduction of NCC does not contribute to the improvement of wastewater treatment performance. When using a membrane with a 5% NCC addition, the treatment efficiency in terms of BOD and COD was 34% and 30%, respectively. The best results were obtained when using a membrane made of polyethersulfone. However, the addition of NCC increased the mechanical and thermal properties of the membrane. In general, in terms of COD and BOD removal efficiency, PMFC is superior to the claimed method of membrane wastewater treatment.

The process of purification for a model aqueous solution containing Cr (VI) is presented in [144]. Kaolin was used as a sorbent for water purification, which was crushed, sieved, and modified (enrichment and calcination). The removal efficiency for chromium depended on its concentration, pH, amount of sorbent, and temperature. The highest degree of Cr (VI) removal was recorded at a pH value of 10 and at a low concentration (70 mg/dm^3). An increase in chromium removal efficiency up to 84% was observed at a kaolin sample weight of 3 g. In general, this method has approximately similar efficiency in chromium removal in comparison with PMFC.

In [145], a method of the secondary biological treatment of wastewater from a poultry processing plant using the microalgae *Chlorella vulgaris* was used. The wastewater was used at different dilutions (25%, 50%, 75%, and 100%). The results showed that increasing the concentration of wastewater leads to the intensive development of the algae biomass, but there is a decrease in COD treatment efficiency (up to 88% when wastewater was used without dilution). In general, this method exceeds PMFC systems in terms of BOD_5 and COD by 25% and 13%, respectively. However, the efficiency of treatment for total nitrogen in the PMFC system was exceeded by almost 30%.

In [146], immobilized bacteria and *Caulerpa lentillifera* microalgae were used for water purification. The biofilter was obtained by immobilizing bacteria onto a chitosan-based aerogel material. The results showed that the simultaneous use of bacteria and microalgae provided water treatment efficiencies of up to 69%, 59%, and 34% for COD, total nitrogen, and total phosphorus. At the same time, the efficiency of wastewater treatment using PMFC is not inferior to this method.

In [147], activated sludge dominated by *Pseudomonas medocina* and *Bacillus* bacteria was used to treat wastewater from SDS. The results showed that the efficiency of wastewater treatment from surfactants was almost 100%, while the treatment rate depended on the surfactant concentration and the amount of inoculum. The degree of purification with activated sludge exceeded the parameters for the PMFC system.

Thus, the efficiency of wastewater treatment for COD and total nitrogen using biological treatment methods practically does not differ from PMFC. Physical methods (membrane filtration and adsorption on kaolin) also do not significantly exceed the PMFC system in wastewater treatment efficiency in terms of COD, nitrogen, and Cr (VI). The physicochemical method (electrooxidation and electrocoagulation) is superior to biological wastewater treatment methods in terms of the rate and may be more preferable for nitrate removal from water. Nevertheless, PMFC systems imply obtaining not only plant phytomass, which can be used for processing into useful products, but also electricity, which distinguishes this technology from biological and physical methods.

3.4. Integration of PMFC into Hydrobotanical Sites for Wastewater Treatment as a Prospect for Further Development of Bioelectric Systems

The PMFC design features, organized according to the "constructed-wetland" technology, can replace artificial wetlands, which have been used for more than 50 years [148] and are intended for wastewater treatment: household wastewater [149], wastewater from fisheries farms [150], and municipal–domestic wastewater from small settlements [151]. After wastewater treatment, their reuse is possible [151,152].

The artificial constructed-wetlands (CW) operation principle is based on natural wastewater treatment processes when plants macrophytes and microorganisms are used, while purification is carried out under controlled conditions [148]. Wastewater can be purified from biogenic elements, heavy metals, and organic pollutants, including surfactants in such systems.

There is a well-known work on the use of CW laboratory models for the removal of synthetic surfactants from the wastewater of car washes [153]. The results show that the efficiency of SDS removal (the main component of the detergent in this study) is 90% when loading 75 dm^3/m^2 and when using CW with the Phragmites australis plant.

The work [154] investigated the removal degree of SDS, polyethylene glycol (PEG), and trimethylamine (TMA) using the plant Phragmites australis. The results show that the greatest removal degree of the selected model toxicants occurs within 7 days, and then sorption is significantly reduced. Additionally, the plants show signs of chronic toxicity over a long period of time. The toxicants have been found to accumulate in various plant components, which makes them unsuitable for use as feed for cattle. The removal degree of SDS is 35% in 35 days.

The integration of PMFC into hydro-mechanical sites is a promising area of scientific research [155]. The use of PMFC makes it possible to achieve more efficient (by 30–50%) wastewater treatment compared to common CW [156]. However, the number of studies on the large-scale application of PMFC is limited [157]. It is necessary to solve a number of problems to scale PMFC systems in the future: high internal resistance [158,159], the selection of the optimal association of microorganisms to reduce competition between them [159], and the biofouling of the cathode, which worsens the diffusion of oxygen to its surface [160,161].

Indeed, the internal resistance (R_{int}) of a PMFC is a combined value consisting of anode and cathode overvoltage and ohmic losses, which are related to the resistance to charge transfer [31]. The internal resistance negatively affects the electrochemical parameters of the PMFC [130,162] and can be decreased by reducing the distance between the electrodes [163], using more efficient proton exchange membranes (if applied) [81]. Bacteria-producing electron transport mediators [164] can be used as an approach to reduce anodic overvoltage. Riboflavins [165], phenazines [166–168], pyocyanins [169], and quinones [170] are distinguished among endogenous media. It should be noted that the use of a mediator increases the current density by increasing the electron transfer rate [166,171]. Various methods of anode [172,173] and cathode [174,175] modification are effective ways to reduce R_{int} (improving the conductivity of the material and reducing the charge transfer resistance). However, such approaches are most often used for MFC, so methods for improving the properties of electrode materials are of interest for implementation in PMFC.

The problem of cathode biofouling can arise during the long-term operation of PMFC. The biofouling process is influenced by the material, surface roughness and charge, and ionic strength of the electrolyte [176]. In general, biofouling leads to a decrease in the oxygen transfer to the catalyst layer, increases the charge transfer resistance, and impairs the proton transfer to the cathode, which leads to an increase in the internal resistance of the devices [161]. Microbial separators [177], modifications with nanoparticles that have catalytic oxygen reduction and antimicrobial activity [178], the use of innovative designs to enable rapid cathode replacement [179], cathode treatment using an alternating current [180], the use of graphene oxide [181], and the use of antibiotics as part of the cathode catalyst [182] have been employed to reduce the effects of this process. Despite the numerous methods used to control biofouling, the most effective strategies are to develop bifunctional cathode catalysts that will simultaneously increase the generated power of PMFC systems, reduce internal resistance, and prevent microbial growth on the cathode without requiring frequent replacement.

It is essential to focus on improving the efficiency of wastewater treatment systems and generating electricity, which is achieved with a detailed examination of bioelectrochemical processes that occur in BES systems.

4. Conclusions

The study of the application possibilities and the development of effective PMFC technologies is relevant to creating new renewable energy sources and is important for solving environmental problems. A PMFC system has autonomy and is able to purify soil and wastewater ecosystems from a wide range of organic and inorganic compounds due to the influence of plants and microorganisms on each other. Thus, the review shows that the efficiency of soil purification from heavy metals with the help of PMFC is, on average, about 30% and 90% from oil. The reduction of such indicators as BOD and COD for wastewater reaches 71% and 52%, respectively. A decrease in biogenic elements in the form of phosphates and nitrates is observed when a PMFC is exploited. Purified water can be reused for household needs. The electricity generated via a PMFC is ecologically clean and can be used to power and charge low-power devices. Moreover, the system can be used as a biosensor in monitoring plant conditions. Decentralized electricity generation and the low cost of manufacturing PMFCs can be used in developing countries, and the generation of electricity in wetlands can become another application of PMFCs.

The problems of operation, which include high resistance and low power, can be solved through the careful choice of the material for the anode and cathode manufacturing by modifying the electrodes in order to both improve their electroconductive properties and increase the electron transfer rate. Moreover, it is proposed to focus on plants with C_4-type photosynthesis, which is more efficient compared to C_3 photosynthesis. More attention should be paid to the selection of the most effective exoelectrogenic types of bacteria to increase a PMFC system's power.

This review examined studies on biological wastewater treatment for synthetic surfactants. Bacteria of the genus *Pseudomonas* sp. are promising bacteria capable of biodegradation and are most popular for household purposes involving the group of anionic surfactants. Some species of these bacteria are capable of producing compounds of the phenazine series. They have been previously used in MFCs. Therefore, this genus of bacteria has the potential for use in a PMFC system, which will combine wastewater treatment from surfactants and electricity generation.

Thus, the possibility of wastewater treatment using PMFC technology with its reuse for household purposes, reduction of greenhouse gases, and low amount of waste during operation favorably distinguishes it from traditional alternative sources of electricity despite the low power output.

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